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Is it possible to induce a fast de-excitation of the 16^+ isomeric state in ^{178}Hf?

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Abstract

The 16^+ level of ^{178}Hf at 2.446 MeV is interpreted as a four-quasi-particle state with K = 16. It decays mainly to a 13^- level at 2.433 MeV, a member of rotational band built on a two-quasi-particle 8^- state (K = 8) at 1.1474 MeV. The 13-keV transition is > 99% E3 character and occurs predominantly through internal conversion. The five-times K-forbidden E3 transition has a large hindrance factor; the half-life of the 16^+ state is known to be 31 years.

If this isomeric nucleus can be induced to release its energy quickly, the resulting energy release would be 6 orders of magnitude more energetic per reaction than that for existing high explosives.

The following mechanisms to enhance de-excitation of this isomer are presented for discussion:

1. inelastic scattering of high-energy neutrons;

2. inelastic scattering of a high-energy, high-intensity electron beam; and

3. interaction with intense photon fields (rf, laser, x ray, γ ray).

Experiments to explore such de-excitation mechanisms are being discussed.

Introduction

In "Table of Isotopes" (7th edition) by Lederer and Shirley[1] there are about 24 excited nuclei with half-lives longer than a month. The excitation energies vary from a few keV to a few MeV.

An example of a promising isomer is the 31-yr half-life excited state of ^{178}Hf with an excitation energy of 2.446 MeV. Macroscopic quantities of this isomer have been created in the LAMPF beam stop.[2] It can also be created by neutron capture of ^{177}Hf (a stable isotope with relative abundance of 18.6%) with a production cross section of $(2 \pm 1) \times 10^{-7}$ barns.[3]

In evaluating possible exploitation of these energetic nuclei, the central questions are

1. What are possible mechanisms of inducing rapid de-excitation?

2. How muc is the natural nuclear decay rate enhanced?

We first review existing information for the ^{178}Hf nucleus. The detailed description that follows indicates that the gross features of the nucleus can be understood with existing nuclear models. We then discuss several schemes for inducing rapid de-excitation. We close by considering experiments that could be attempted in Los Alamos.

The Structure of the ^{178}Hf Nucleus

The high-spin isomeric state was first reported by Helmer and Reich.[4] Subsequently, several detailed studies[3,5-9] have been reported. The partial level scheme populated by the decay of the isomeric state is shown in Fig. 1. Combined with additional information,[1,10] about sixty levels exist below 3 MeV with reasonably well-known spin and parities. Almost all of these levels can be fitted as members of different rotational bands with standard formulas:

$$E(I) = C_1 + C_2 I(I+1) + C_3 I^2(I+1)^2, \quad (K \neq 1) \text{ and}$$

$$E(I) = C_1 + C_2 I(I+1) + C_3 I^2(I+1)^2 + (-1)^{(I+1)} I(I+1)(C_4 + C_5 I(I+1)),$$

$(K=1)$.

The fitted results are plotted in Fig. 2. Many rotational band heads can be considered as two- or four-quasi-particle states. The relative positions of single-particle orbitals are shown in Fig. 3.[4]

The ground state is assigned[3] as $(^7\!/_2[404]_p)^2_{0+}$, $(^7\!/_2[514]_n)^2_{0+}$. The two 8^- states at 1147.4 and 1479.0 keV are considered as mixing of two-quasi-particle states from the

$$\{^9\!/_2[624]_n + {}^7\!/_2[514]_n\}_{8-} \text{ and } \{^7\!/_2[404]_p + {}^9\!/_2[514]_p\}_{8-}$$

configurations.[4] The two 1^- states at 1310.0 and 1513.7 keV may be considered as mixing of the same two-quasi-particle configurations aligned in the opposite direction, that is the mixing of $\{^9\!/_2[624]_n - {}^7\!/_2[514]_n\}_{1-}$ and $\{^9\!/_2[514]_p - {}^7\!/_2[404]_p\}_{1-}$. The 2^- state at 1260.5 keV is considered as the two-quasi-particle state $\{^5\!/_2[512]_n - {}^9\!/_2[624]_n\}_{2-}$. The 4^+ state at 1514 keV may be $\{^7\!/_2[514]_n + {}^1\!/_2[510]_n\}_{4+}$ and the 6^+ state at 1554 keV may be $\{^7\!/_2[404]_p + {}^5\!/_2[402]_p\}_{6+}$ (Ref. 7).

The 14^- state at 2573.5 keV, which was first reported by Khoo and Lovhoiden with the ^{176}Yb(α,2n)^{178}Hf reaction,[7] decays to the 16^+ state at 2446 keV with an M2 (126 keV) transition, to the 13^- state at 2433.3 keV with an M1 (140.3 keV) transition, and to the 12^- state at 2136.5 keV with an E2(437.0 keV) transition. The half-life of this 14^- state is 68 μs. This state is considered[7] as a four-quasi-particle state with configurations $\{^9\!/_2[624]_n + {}^7\!/_2[514]_n + {}^7\!/_2[404]_p + {}^5\!/_2[402]_p\}_{14-}$.

The level energy of the 16^+ state was determined to be 2446 keV with internal conversion lines associated with a weak M4 (309.50 keV) transition to the 12^- state at 2136.5 keV;[8] the 16^+ state decays predominantly to the 13^- state at 2433.3 keV. This 12.7-keV transition is > 99% E3, and occurring mainly by internal conversion. The five-times K-forbidden E3 transition has a large hindrance factor; the half-life of the 16^+ state is 31 yr. This state is considered as four-quasi-particle state with configuration $\{9/2[624]_n + 7/2[514]_n + 7/2[404]_p + 9/2[514]_p\}_{16^+}$.

Figure 4 shows the partial decay scheme of ^{176}Yb(α,2n)^{178}Hf reaction.[7]

If the 16^+ level energy is determined by the M4 309.5-keV transition ($16^+ \rightarrow 12^-$) to be 2446.0 keV,[8] then the transition between 14^- to 16^+ would be 127.5 keV as compared with 126.1 keV observed in (α,2n) reactions of Khoo and Løvhøiden.[7]

The interband transitions from the upper 8^- band directly to the lower 8^- band appear to dominate any intraband transitions. While such behavior provides a signature of mixing between the bands, the absence of the intraband transition in the upper 8^- band requires explanation, which is not offered by Khoo and Løvhøiden.[7]

In a Coulomb excitation study, Hamilton <u>et. al.</u>[9] found that the lower $K^\pi = 8^-$ isomer ($T_{1/2} = 4$ s) was populated. The mechanism for the population is not understood. Hypothetically there exists a state connected to the ground-state band by an E2 transition. It is possible for this state to be excited via multiple Coulomb excitation and yet have a strong coupling directly (or perhaps through an intermediate state) to the lower $K^\pi = 8^-$ band.[9]

The half-lives of other possible modes of decay have been estimated[8] for

1. β-decay to high-spin states of ^{178}Ta; $T_{1/2} \sim 10^4$ yr;

2. electron capture to high-spin states of ^{178}Lu; $T_{1/2} \sim 3 \times 10^{3}$ yr;

3. α decay to ^{174}Yb; $T_{1/2} \geq 6 \times 10^{8}$ yr; and

4. spontaneous fission, $T_{1/2} \sim 10^{9}$ yr.

Possible Mechanism to Induce Fast De-Excitation

I. Neutron inelastic scattering; ^{178m_2}Hf(n,n γ)^{178}Hf

The fact that the 16^{+} isomeric state can be produced by neutron capture of ^{177}Hf indicates that the incoming neutron can induce changes in single-particle orbitals.

Table 1 lists cross sections for the (n,γ) population of various states in ^{178}Hf. To populate the 8^{-} states, the incoming neutron must convey four units of angular momentum to populate neutron orbital $^{9}/_{2}$[624] or to excite one proton to an orbital of $^{9}/_{2}$[514]. The difference in cross section leading to the two 8^{-} levels can be understood in terms of different mixing. To populate the 16^{+} state, the incoming neutron must be in the higher neutron orbital $^{9}/_{2}$[624], as well as excite one proton from $^{7}/_{2}$[404] to $^{9}/_{2}$[514].

With neutron inelastic scattering, the following cases will lead to fast de-excitation:

1. Excitation of the proton from the $^{9}/_{2}$[514] orbital to $^{5}/_{2}$[402] results in population of the 14^{-} state, which has a 68-µs half-life.

2. The two $K^{\pi} = 1^{-}$ bands, which can be considered as the coupling of the same neutron and proton orbitals as the $K^{\pi} = 8^{-}$ bands, but aligned oppositely, have levels located less than 200 keV above $K^{\pi} = 8^{-}$ bands. It is quite possible that the $K^{\pi} = 9^{+}$ and 7^{+} bands based on different alignment of four-quasi-particle $^{9}/_{2}[624]_{n}$, $^{7}/_{2}[514]_{n}$, $^{7}/_{2}[404]_{p}$ and $^{9}/_{2}[514]_{p}$ are not too far above the 16^{+} state. Levels in both the 9^{+} and 7^{+} bands should have short half-lives.

3. The $^1/_2[541]$ proton orbital lies between the $^9/_2[514]$ and $^5/_2[402]$ orbitals. A four-quasi-particle state with I, K^π = 12, 12^+, based on $\{^7/_2[514]_n + {}^9/_2[624]_n + {}^7/_2[404]_p + {}^1/_2[541]_p\}_{12^+}$, may exist between the 16^+ and 14^- levels. With its lower K value, it should decay rapidly to the lower 8^- band.

II. Interaction with Intense Photon Fields

1. With the 14^- level only 126 keV above the 16^+ level, the nucleus may be excitable from 16^+ to 14^- by photon (gamma-ray) resonance absorption.

2. Enhancement of nuclear β-decay with laser and rf fields, which has been proposed by several investigators[11] should also apply to the internal conversion process. The decay of the 16^+ to the 13^- and 12^- levels by internal conversion should be enhanced when $^{178m_2}Hf$ nuclei are exposed to intense rf/laser/synchrotron radiation fields. Such exposure may also enhance the β-decay to ^{178}Ta or electron capture to ^{178}Lu. Present evidence indicates, however, that the enhancement factor may be very small.[11]

III. Interaction with a High-Energy, High-Current Electron Beam

It is not difficult to find high-energy (~ 20 MeV), high-current (~ 1 kA) electron beams for application to the following two cases:

1. Electron inelastic scattering may change single-particle orbitals, like for neutron inelastic scattering, to lower K levels that will decay rapidly.

2. The first few nuclear excited states can be populated due to a thermal equilibrium effect.[12] When the 16^+ isomer is exposed in high-power electron beams, many levels above 16^+ may become populated, and these levels could decay rapidly.

Proposed Experiments

At present, theoretical knowledge and computational capability are inadequate to predict the enhancement of de-excitation; therefore, experiments are proposed to establish any decay-rate enhancement.

The following facilities at Los Alamos (or facilities elsewhere) can be used:

1. PSR-WNR for neutron inelastic scattering experiments.

2. The PHERMEX facility for electron beam, rf field, and gamma-ray (bremsstrahlung) experiments.

3. CO_2 or KrF lasers for laser interaction experiments.

The experimental techniques are common to all these laboratory experiments. Regardless which mechanism induces fast decay, the intraband transitions in the lower 8^- band provide the signature (except for β-decay and electron-capture). The ideal set-up will include several gamma-ray detectors placed at different angles with respect to the beam line. Single gamma spectra and coincidences between any pair of detectors will be recorded with multichannel analyzers. Increases in intraband transition intensities imply that fast de-excitation has been induced. The coincidence spectra will yield additional information of the energies of new levels excited, and the angular distribution of new gamma rays or $\gamma\gamma(\theta)$ with known gamma rays may be used to determine the spins and parities of these new levels.

Conclusions

1. We have enough information to understand the structure of the ^{178}Hf nucleus and to propose mechanisms for inducing rapid de-excitation. However, the existing nuclear theory is inadequate to predict microscopic behavior such as the exact locations of proton and neutron orbitals and how they couple, reaction cross-sections, and decay rate enhancement.

2. We have access to macroscopic quantities of $^{178m_2}Hf$, and several facilities in Los Alamos are available for decay-rate enhancement experiments.

3. The proposed experiments will determine whether or not highly enhanced decay rates of excited states can be achieved. If enhanced decay rates are found, a new source of very high specific energy yield material will be available.

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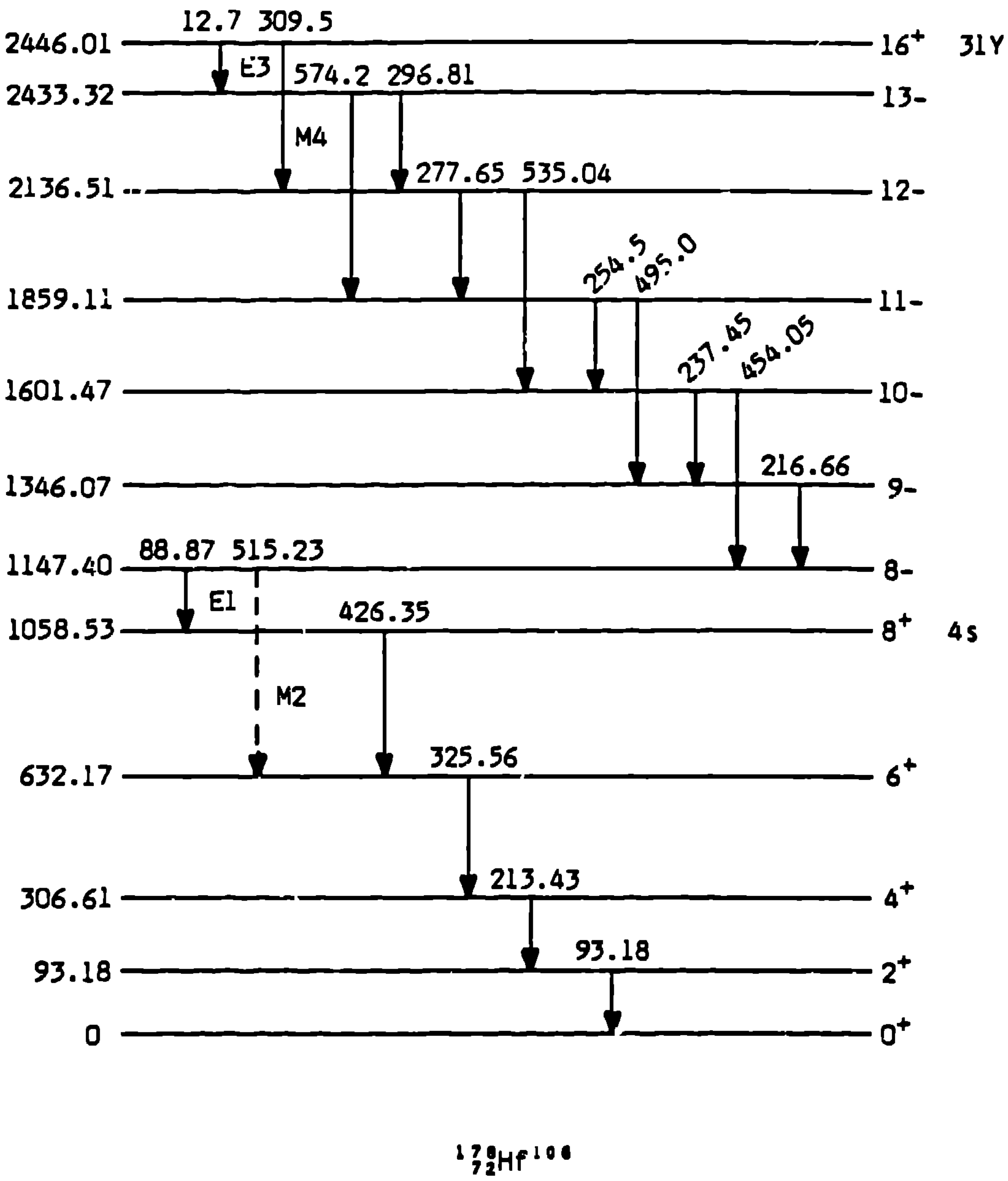

Fig. 1. Decay scheme of 16^+ isomer state in $^{178}_{72}Hf$.

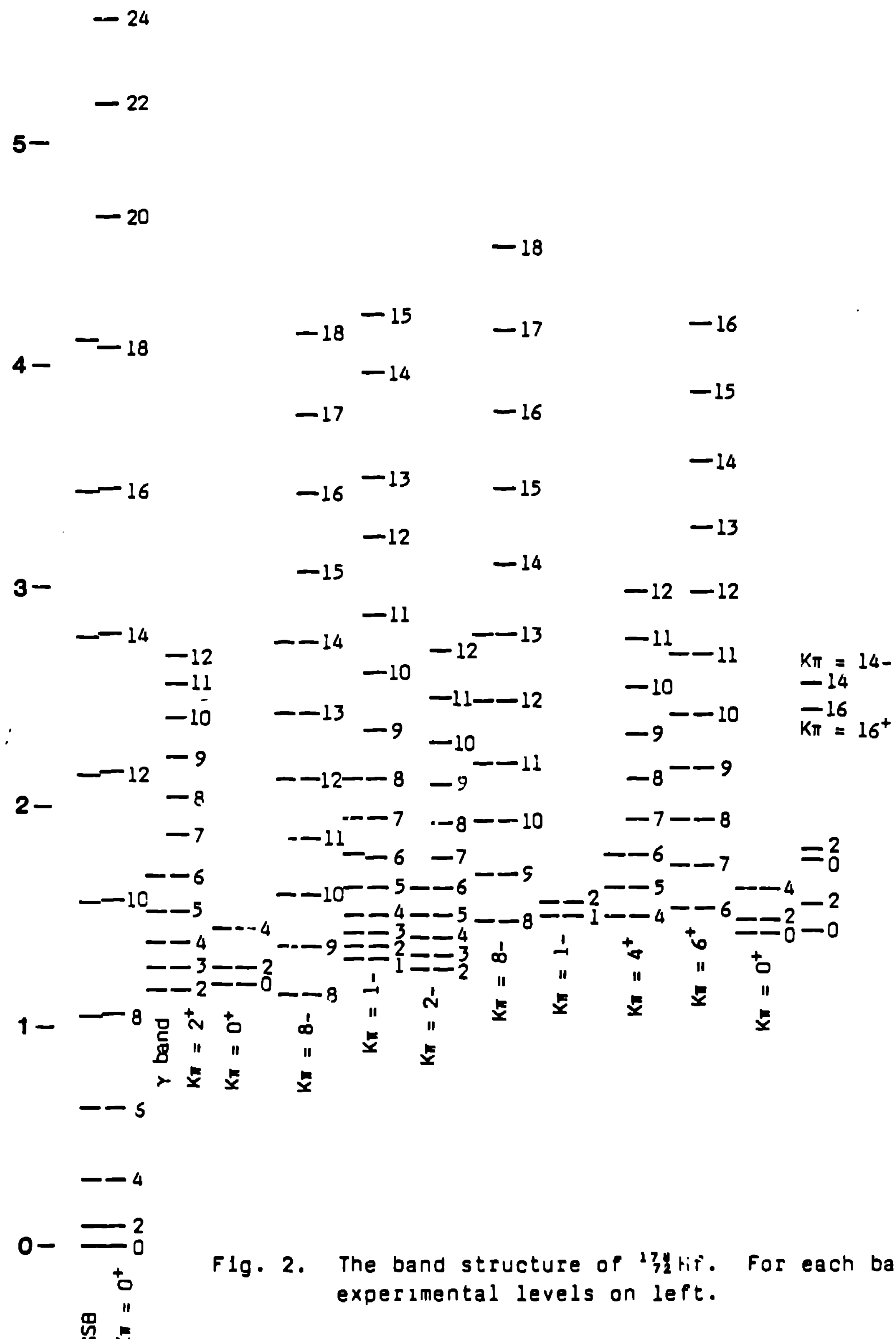

Fig. 2. The band structure of $^{178}_{72}$Hf. For each band, experimental levels on left.

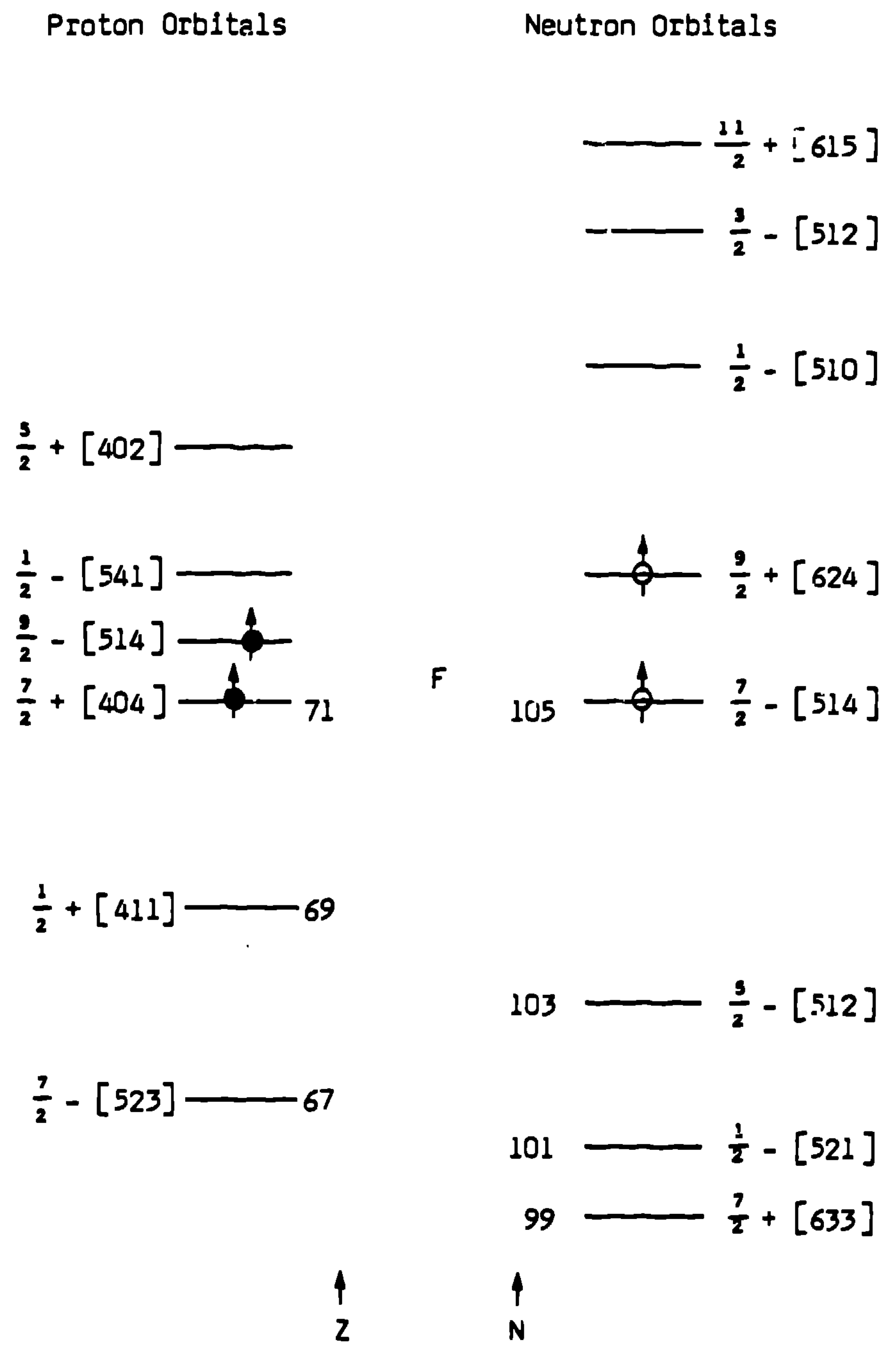

Fig. 3. Relative positions of the single-particle orbitals in region of $Z = 72$ and $N = 106$. The Fermi surface for the neutron and proton system in $^{178}_{72}Hf$ is indicated by the symbol F.

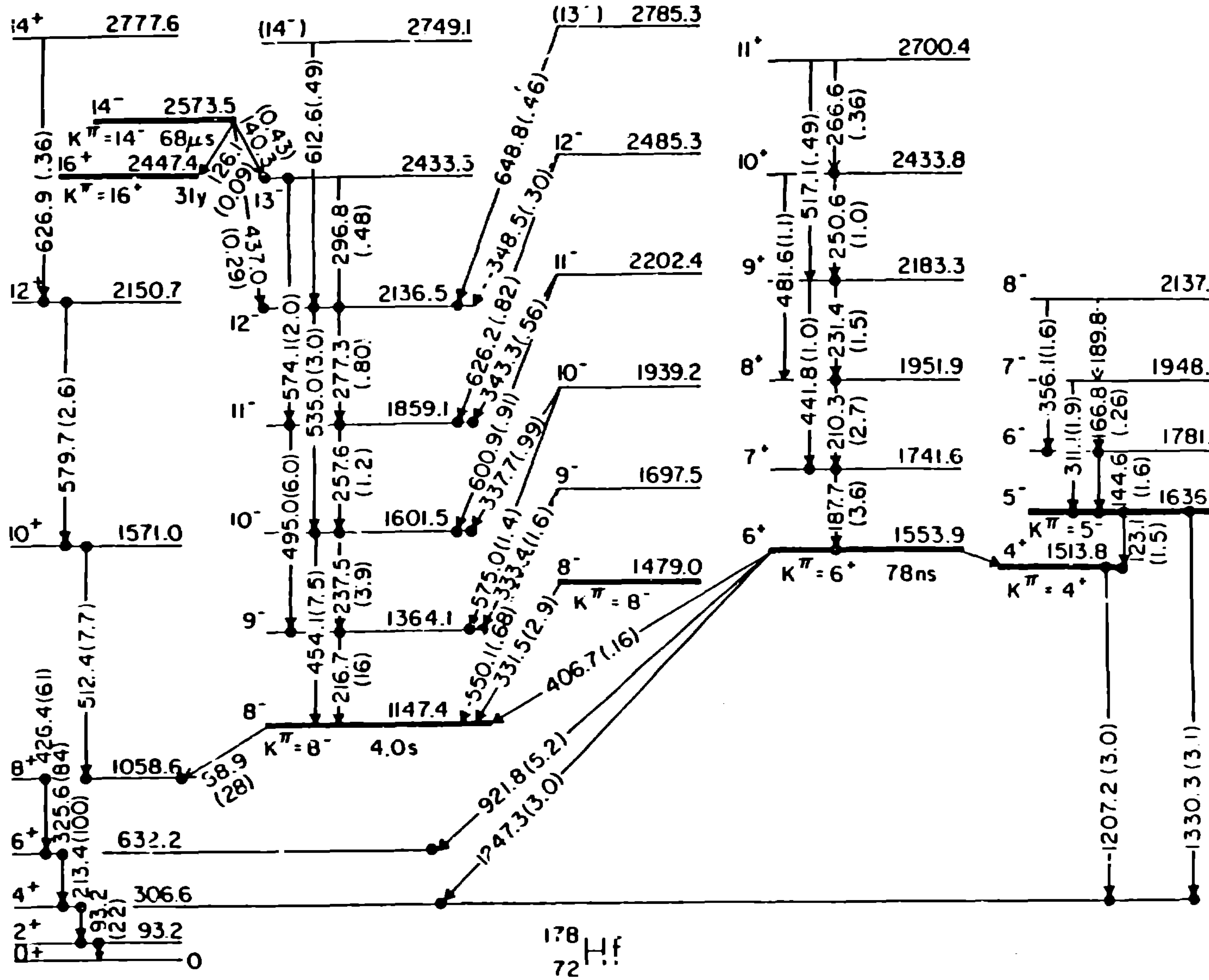

Fig. 4. Partial decay scheme of ^{178}Hf, from Ref. 7, with K ≥ 4, decay branches of IK = 6 6 and 4 4 band heads proceeding through the gamma-band are not shown, relative gamma-ray intensities in ^{176}Yb(α,2n)^{178}Hf reaction at 26 MeV α energy are given in brackets.

Table 1. Cross sections for (n,γ) population of various states in ^{178}Hf (see Ref. 3 and 9).

State	E(keV)	I^{π}	Single-particle orbitals	Cross Section (barn)
Initial	0	0^{7}	$\left\{(\tfrac{7}{2}[404]_p)^2_0 + \tfrac{7}{2}[514]_n\right\}_{7/2^-}$	
Final	0	0^+	$\left\{(\tfrac{7}{2}[404]_p)^2_0 + (\tfrac{7}{2}[514]_n)^2_0\right\}_{0^+}$	365
	1147	8^-	$36\% \left\{(\tfrac{7}{2}[404]_p + \tfrac{9}{2}[514]_p)_{8^-} + (\tfrac{7}{2}[514]_n)^2_0\right\}_8$ $+64\% \left\{(\tfrac{7}{2}[404]_p)^2_0 + (\tfrac{7}{2}[514]_n + \tfrac{9}{2}[624]_n)_{8^-}\right\}_{8^-}$	0.93
	1479	8^-	$64\% \left\{(\tfrac{7}{2}[404]_p + \tfrac{9}{2}[514]_p)_{8^-} + (\tfrac{7}{2}[514]_n)^2_0\right\}_{8^-}$ $+36\% \left\{(\tfrac{7}{2}[404]_p)^2_0 + (\tfrac{7}{2}[514]_n + \tfrac{9}{2}[624]_n)_{8^-}\right\}_{8^-}$	0.25
	2446	16^+	$\left\{\tfrac{7}{2}[404]_p + \tfrac{9}{2}[514]_p + \tfrac{7}{2}[514]_n + \tfrac{9}{2}[624]_n\right\}_{16^+}$	$(2 \pm 1) \times 10^{-7}$